

PCT

NOTIFICATION OF ELECTION

(PCT Rule 61.2)

From the INTERNATIONAL BUREAU

To:

Commissioner
US Department of Commerce
United States Patent and Trademark
Office, PCT
2011 South Clark Place Room
CP2/5C24
Arlington, VA 22202
ETATS-UNIS D'AMERIQUE
in its capacity as elected Office

Date of mailing:

03 May 2001 (03.05.01)

International application No.:

PCT/GB00/03943

Applicant's or agent's file reference:

15438 LgCm

International filing date:

16 October 2000 (16.10.00)

Priority date:

22 October 1999 (22.10.99)

Applicant:

RAYBONE, David et al

1. The designated Office is hereby notified of its election made:



in the demand filed with the International preliminary Examining Authority on:

02 February 2001 (02.02.01)



in a notice effecting later election filed with the International Bureau on:

2. The election ☒ was

was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO
34, chemin des Colombettes
1211 Geneva 20, Switzerland

Facsimile No.: (41-22) 740.14.35

Authorized officer:

J. Zahra

Telephone No.: (41-22) 338.83.38

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From the INTERNATIONAL BUREAU

**NOTIFICATION OF THE RECORDING
OF A CHANGE**

(PCT Rule 92bis.1 and
Administrative Instructions, Section 422)

To:

LOFTING, Marcus, John
Accentus plc
Patents Department
329 Harwell
Didcot
Oxfordshire OX11 0QJ
ROYAUME-UNI

Date of mailing (day/month/year)

26 July 2001 (26.07.01)

Applicant's or agent's file reference

15438 LgCm

IMPORTANT NOTIFICATION

International application No.

PCT/GB00/03943

International filing date (day/month/year)

16 October 2000 (16.10.00)

1. The following indications appeared on record concerning:



the applicant



the inventor



the agent



the common representative

Name and Address

AEA TECHNOLOGY PLC
329 Harwell
Didcot
Oxfordshire OX011 0QJ
United Kingdom

State of Nationality

GB

State of Residence

GB

Telephone No.

Facsimile No.

Teleprinter No.

2. The International Bureau hereby notifies the applicant that the following change has been recorded concerning:



the person



the name



the address



the nationality



the residence

Name and Address

ACCENTUS PLC
329 Harwell
Didcot
Oxfordshire OX011 0QJ
United Kingdom

State of Nationality

GB

State of Residence

GB

Telephone No.

Facsimile No.

Teleprinter No.

3. Further observations, if necessary:

4. A copy of this notification has been sent to:



the receiving Office



the International Searching Authority



the International Preliminary Examining Authority



the designated Offices concerned



the elected Offices concerned



other:

The International Bureau of WIPO
34, chemin des Colombettes
1211 Geneva 20, Switzerland

Authorized officer

S. Buttay

Facsimile No.: (41-22) 740.14.35

Telephone No.: (41-22) 338.83.38

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REC'D 31 JAN 2002

WIPO PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference 15438 LgCm	FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/GB00/03943	International filing date (day/month/year) 16/10/2000	Priority date (day/month/year) 22/10/1999
International Patent Classification (IPC) or national classification and IPC B01D53/32		
Applicant [AEA TECHNOLOGY PLC et al.] ACCENTUS PLC et al.		



1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of 7 sheets, including this cover sheet.
☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of 15 sheets.

**CORRECTED
VERSION**

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☒ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☒ Certain defects in the international application
- VIII ☐ Certain observations on the international application

Date of submission of the demand 02/02/2001	Date of completion of this report 29.01.2002
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer Connor, M Telephone No. +49 89 2399 8402 

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT**

International application No. PCT/GB00/03943

I. Basis of the report

1. With regard to the **elements** of the international application (*Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17)*):

Description, pages:

2,7,9-14,16	as originally filed	
1,1a,3-5,5a,6,8, 15,15a	with telefax of	16/01/2002

Claims, No.:

21,22	as originally filed	
1-20	with telefax of	16/01/2002

Drawings, sheets:

1/1	as originally filed
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2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT**

International application No. PCT/GB00/03943

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:
- ☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

III. Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

1. The questions whether the claimed invention appears to be novel, to involve an inventive step (to be non-obvious), or to be industrially applicable have not been examined in respect of:

- ☐ the entire international application.
- ☒ claims Nos. 2-15, 17-20.

because:

- ☐ the said international application, or the said claims Nos. relate to the following subject matter which does not require an international preliminary examination (*specify*):
 - ☐ the description, claims or drawings (*indicate particular elements below*) or said claims Nos. are so unclear that no meaningful opinion could be formed (*specify*):
 - ☒ the claims, or said claims Nos. are so inadequately supported by the description that no meaningful opinion could be formed.
 - ☐ no international search report has been established for the said claims Nos. .
2. A meaningful international preliminary examination cannot be carried out due to the failure of the nucleotide and/or amino acid sequence listing to comply with the standard provided for in Annex C of the Administrative Instructions:
- ☐ the written form has not been furnished or does not comply with the standard.
 - ☐ the computer readable form has not been furnished or does not comply with the standard.

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability;

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT**

International application No. PCT/GB00/03943

citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes: Claims
	No: Claims 1,16
Inventive step (IS)	Yes: Claims
	No: Claims 1,16
Industrial applicability (IA)	Yes: Claims 1-20
	No: Claims

**2. Citations and explanations
see separate sheet**

VII. Certain defects in the international application

The following defects in the form or contents of the international application have been noted:
see separate sheet

Re Item I

Basis of the report and

Re Item III

Non-establishment of opinion with regard to novelty, inventive step and industrial applicability

1.1 As the applicant did not indicate neither the location of, nor the basis for the extensive amendments filed with its letter dated 16.01.2002, compliance to Article 34(2)(b) PCT was examined for the amended independent claims 1 and 16 only. The amendments of the description and dependent claims was not examined.

In a national or regional phase, the applicant may be requested to **clearly** and **explicitly** indicate the location of, and basis for the filed amendments in order to allow examination to proceed conveniently.

1.2 Amended claims 1 and 16 are considered to comply with the requirements of Article 34(2)(b) PCT.

Re Item V

Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1 Reference is made to the following documents:

- D1: US-A-5 746 984 (HOARD JOHN W) 5 May 1998 (1998-05-05)
- D2: PATENT ABSTRACTS OF JAPAN vol. 1999, no. 09, 30 July 1999 (1999-07-30) & JP 11 114351 A (SANDENSHA:KK;YAMAMOTO TOSHIAKI; YOU SADAYOSHI), 27 April 1999 (1999-04-27)
- D3: US-A-5 715 677 (VOGTLIN GEORGE E ET AL) 10 February 1998 (1998-02-10)
- D4: PATENT ABSTRACTS OF JAPAN vol. 1999, no. 09, 30 July 1999 (1999-07-30) & JP 11 114359 A (AGENCY OF IND SCIENCE &TECHNOL), 27 April 1999 (1999-04-27)

2 Before proceeding to the substantive examination of the subject matter of claims 1 and 16, some comments concerning their respective wording is required.

2.1 The wording of claim 1 defines that carbonaceous particulates including soot are trapped in the active bed (320), but does not exclude that any other components of the exhaust gas, e.g., NO_x, may be trapped too.

- 2.2 The expression in claim 1 (l. 23) "is much less likely to occur" is not clear in the sense of Article 6 PCT, because vague and indefinite.
- 2.3 The characterizing part of claim 16 is drafted in terms of functional features referring to the use of the claimed apparatus and not in terms of technical features as required by Rule 6.3(a) PCT.
- 3 The method proposed in **claim 1** and the product in **claim 16** of the present application cannot be considered as novel (Article 33(2) PCT) for the following reasons.
- 3.1 D3 discloses a method of treating gases comprising a mixture of NO_x and particulate emissions (cf. D3, Abstract: "particulates from diesel engines" invariably comprise soot and hydrocarbons) according to the preamble of claim 1 of the present application (cf. point V-3 of the first communication dated 23.10.2001) characterized in that electrical potential is applied to generate a non-thermal plasma (cf. D3, col. 2, l. 1) whereby trapped species have a longer residence time than non trapped ones (rather obvious), and the trapped species include the carbonaceous particulates including soot (cf. D3, abstract: "physical trap for the particulate matter"). Said carbonaceous particulates including soot are air oxidised (cf. D3, col. 2, l. 2) while conversion of NO to NO_2 is less likely to occur, since NO_x was decomposed into N_2 and O_2 (cf. D3, col. 3, l. 12).
- 3.2 All the features called for in claim 16 of the present application excluding the functional features (cf. point V-2.3 supra) are anticipated in combination by each of D1-D4, as demonstrated in Table 1 of the first communication. Consequently, the reactor *per se* (i.e., independently of its use) of claim 16 cannot be considered as novel in the sense of Article 33(2) PCT.
- 3.3 All the features called for in claim 16, including the functional features are disclosed in each of D1 and D3 (cf. point V-3.1 supra). NO selective catalyst is disclosed in D1, col. 4, l. 6; and in D3, col. 4, l. 13: "bed of granular (solid) absorbent/catalyst material", which is actually the same bed which traps the carbonaceous particulates). Consequently, even with its functional features, the reactor of claim 16 cannot be considered as novel in the sense of Article 33(2) PCT.

Re Item VII

Certain defects in the international application

- 1 For lack of clear indications concerning the amendments of the description filed with the applicant's letter dated 14.01.2002, fulfilment of the requirements of Rule 5.1(a)(iii) PCT was not established. The applicant's attention may be drawn to said Rule in case it would proceed to a national or regional phase.

Reactor for Plasma Treatment of Gases

The invention relates to a method and a reactor for plasma treatment of gases utilising a non-thermal plasma.

5

There is increasing interest in the use of non-thermal plasmas for treatment of gaseous exhausts and in particular for treatment of exhausts from motor vehicles. Treatment of exhausts involves conversion of harmful exhaust components such as hydrocarbons to carbon dioxide and water as well as harmful NO_x components of exhausts to nitrogen. In the case of lean burn engines such as diesels there is a requirement for removal of carbonaceous particulates by for example oxidation to carbon dioxide. Examples of non-thermal treatment of exhausts are described in US 3,983,021 (Monsanto), US 5,147,516 (Tecogen) and US 5,254,231 (Battelle Memorial Institute). GB 2,274,412 (AEA Technology) describes a method for the treatment of diesel emissions by a non-thermal plasma for oxidation of carbonaceous particulates and reduction of NO_x to nitrogen.

US 5,746,984 describes a system in which a storage device collects NO_x , hydrocarbon, or particulate emissions or a mixture of these emissions, during a storage phase while conditions in the gas are oxidising, and destroys the collected emissions in a plasma reactor during a destruction phase while conditions in the gas are non-oxidising. There is indication that it is preferred to maintain an oxidising atmosphere even during the destruction phase, but no indication as to how NO_x can be destroyed in such conditions.

US 5,715,677 describes a system in which NO_x is first absorbed onto a solid absorbent bed that simultaneously acts as a physical trap for particulate matter. The bed is regenerated in a separate, second stage process by
5 pulsed plasma decomposition of NO_x followed by air oxidation of trapped particulate matter. Two beds are used and switched alternately from absorption stage to regeneration phase so that a continuous flow of gas can be dealt with.

10

JP 11114359 and JP 11114351 also disclose systems for adsorbing volatile gaseous constituents on an adsorbant and regenerating the adsorbant in a plasma.

15

Plasma can be used to activate or produce reactant species, which then subsequently react with or without catalytic enhancement to yield the desirable products. For example, our publication WO99/12638 describes the
20 plasma production of plasma activated hydrocarbons as a precursor to the selective catalytic reduction of NO_x to N_2 . Examples of catalysts for this selective reduction of NO_x to nitrogen are alkali metal-exchanged zeolite Y or silver aluminate. Other metal-exchanged or metal doped

constraints, i.e. reactor size, and the acceptable level of power input to the reactor in as far as all of the exhaust passes through the reactor.

5 The present invention is based upon an appreciation of the advantages that follow if one changes the residence time of selected species in the reactor, and thus breaks the, at present unavoidable, link between joules per litre input power and reactant species. This
10 would lead to a simplification in the design and an improvement in the energy efficiency of plasma reactors.

 Examples have been given which show that, without catalytic enhancement, plasma reactors can produce
15 quantities of undesirable by-products usually associated with partial oxidation of hydrocarbons (see 'Analysis of plasma-catalysis for diesel NO_x remediation' by J Hoard and M L Balmer, SAE 982429), for example methyl nitrate, formaldehyde. A solution is offered if hydrocarbons can
20 be retained for relatively long periods of time to achieve complete conversion to CO and CO₂, while oxides of nitrogen may require a short residence time to avoid formation of acids.

25 It is an object of the present invention to provide a method and a non-thermal plasma reactor which addresses these problems.

 The invention provides, in one of its aspects, a
30 method of treating gases which contain nitrogen oxides, carbonaceous particulates including soot, hydrocarbons, and other residual constituents including oxygen, which

method comprises passing the gases through a reactor comprising a bed of active material in an enclosure having gas flow conduits for directing gas flow through or over the bed of active material, applying an
5 electrical potential to generate a non-thermal plasma in gas permeating the active material, at least a component of the active material being such as to adsorb or trap carbonaceous particulates including soot, characterised in that the electrical potential is applied to generate
10 said non-thermal plasma during passage through the active material of the gases undergoing treatment whereby the trapped carbonaceous particulates including soot have a longer effective residence time in the non-thermal plasma relative to species in the gas flow which are not
15 adsorbed or trapped and are oxidised by oxidative species present in the gases while conversion of NO to NO₂ is much less likely to occur.

Preferably the gases are further subjected to the
20 action of a NO selective catalyst, preferably silver doped alumina, which selectively absorbs both NO and hydrocarbons and/or partially oxygenated hydrocarbons and promotes their reaction together to reduce NO directly to N₂.

25

Preferably the gases subjected to the action of a NO selective catalyst are also subjected to further plasma activation which promotes the formation of activated hydrocarbons and/or partially oxygenated hydrocarbons.

30

Preferably the gases are subjected to flow through or over a plurality of beds of active material each of which adsorbs or traps a different predetermined chemical species, and such predetermined chemical species, in

addition to the aforesaid active material for trapping carbonaceous particulates including soot and NO selective catalyst, may be, but is not restricted to, a species from the group comprising nitrogen, oxygen, oxides of

5 carbon such as CO, CO₂, water, hydrocarbons including saturated, unsaturated, cyclic, branched and un-branched hydrocarbons, oxygenated hydrocarbons such as aldehydes, ketones, alcohols, acids ethers and esters, aromatic hydrocarbons and derivatives thereof including poly

10 aromatic hydrocarbon compounds, oil fractions, fuel and partially burned fuel, air and air/fuel mixes, sulphur compounds including SO₂ and sulphates, organo-nitrogen species, acid gases, combustion modifiers/enhancers, additives such as urea, ammonia, cerium oxide (such as

15 Eolys) and plasma activated species such as O, OH, O₃ activated hydrocarbons including partially oxygenated hydrocarbons/ organic molecules and electronically and vibrationally excited state species.

The invention includes a non-thermal plasma reactor for the treatment of gases, which contain nitrogen oxides, carbonaceous particulates including soot, hydrocarbons, and other residual constituents including
5 oxygen, which reactor comprises a bed of active material in an enclosure having gas flow conduits for directing gas to flow through or over the bed of active material, electrodes adapted when electrically energised to generate non-thermal plasma in the gas permeating the
10 active material, at least a component of the active material being such as to adsorb or trap carbonaceous particulates including soot in the gas flow, characterised in that in operation of the reactor said active material increases the effective residence time in
15 the non-thermal plasma of the said carbonaceous particulates including soot relative to the residence time of species in the gas flow which are not adsorbed or trapped, and the trapped carbonaceous particulates including soot are oxidised by oxidative species present
20 in the gases while conversion of NO to NO₂ is much less likely to occur, and an NO selective catalyst is additionally provided for selectively adsorbing both NO and hydrocarbons and/or partially oxygenated hydrocarbons, and promoting their reaction together to
25 reduce NO directly to N₂.

The function of the active material, or component thereof, having the capability to adsorb or trap selected species can be seen as that of a selective filter for
30 that species.

In addition to the advantages, referred to above,

which result from the effective increase in residence time of the selected species, such a selective filter can operate to adsorb or trap the reactants and hold them for sufficient time for them to be activated by a plasma
5 and/or selected filter to a state where they can react with for example NO_x to yield desirable products. In this role the filter material or trapped species in the presence of a plasma can be made to appear to act as a catalytic surface but importantly neither the plasma nor
10 the selective filter nor the trapped species alone need have catalytic properties. Considering the trapped species comprising carbonaceous particulate material from a diesel engine, for example soot that consists mainly of elemental carbon, in the plasma region soot becomes.
15 exposed to plasma generated species for example oxygen atoms. Oxygen atoms or other plasma generated species may diffuse into, adsorb and react with soot. Other plasma generated species include but are not restricted to OH , O_3 and NO_2 . For example it is known oxygen atoms
20 can diffuse into soot and form aldehyde-type groups on the surface. Oxygenated soot has different activated and catalytic properties to non-oxygenated soot. In this example the surface of the reactant material, e.g. carbon is transformed into a catalyst.

25

A selective filter in accordance with the present invention provides, by selective modification of residence times, a method of controlling and hence optimising the distribution of product species from a
30 plasma reactor with a significant degree of independence from flow rate, reactor size or energy density. The method of controlling and hence optimising the distribution of product species may include a method for optimising the desorption of the selectively filtered

assisted processing of the exhaust emissions from internal combustion engines to remove noxious components therefrom consists of a reactor chamber 300 which has inlet and outlet stubs 301, 302, respectively, by means of which it can be incorporated into the exhaust system of an internal combustion engine.

Inside the reactor chamber 300 there is an inner electrode 303 which is supported within a dielectric tube 304, made for example out of γ -alumina which has its upstream end closed by a spherical dome 305 to facilitate the flow of exhaust gases through the reactor. The inner electrode 303 is supported in the dielectric tube 304 by two spider supports 306, 307. The inner surface of the dielectric tube can be metallised with a metal coating in order to increase the physical contact between the electrode and dielectric tube. The support 307 is connected to a high voltage input terminal 308 via a ceramic insulated feed 309 so that a potential of the order of kilovolts to tens of kilovolts and repetition frequencies in the range 50 to 5000 Hz can be applied to the inner electrode 303. Concentric with the inner electrode 303 and dielectric tube 304 is a grounded outer electrode 310 made for example of stainless steel. The dielectric tube 304 and outer electrode 310 are supported within the reactor chamber 300 by disks 311, 312 made of an insulating ceramic material, such as alumina. A compliant heat resistant material 313 is interposed between the electrode support 311 and the dielectric tube 304.

The space between the dielectric tube 304 and the outer electrode 310 is filled with a bed of active material 320, shown in Figure 1, but omitted from Figure 2 for clarity of representation of the gas flow paths.

by activation of hydrocarbons in the exhaust gas stream by for example plasma activation, to form species such as partially oxygenated hydrocarbons, for example formaldehyde (CH_2O). The presence of the non-thermal

5 plasma is important for this in that it is effective for the required activation of hydrocarbons at significantly lower temperatures than those required for thermal production of oxygenated hydrocarbons.

10 It will be apparent to those skilled in the art that this selective filter approach may be applied to other chemical processes, gases and exhaust streams, although these are not the subject of the present invention as claimed, except in so far as they may be applied in
15 combination therewith.

The filter material may be selected for its ability to trap or adsorb a predetermined species in the destruction of toxic waste compounds for example those
20 used in applications such as the micro-electronics and semi-conductor industries. Examples of these include species such as volatile organic compounds, halogen-containing compounds including perfluorocarbons, hydrofluorocarbons and Freons. Increasing the residence
25 time of these species in the plasma by using a selective filter may result in an increase in the efficiency of their destruction.

In another example the selective filter material may
30 be chosen for its ability to trap or adsorb a predetermined species produced in the plasma to coat or modify the filter material in some way.

The filter material may be selected for its ability
35 to trap or adsorb a predetermined species produced in the plasma to prevent a certain gas phase chemical reaction

occurring. This could be to prevent a pollutant or toxin being formed or to act as an inhibitor for a certain

Claims

1. A method of treating gases which contain nitrogen
oxides, carbonaceous particulates including soot,
5 hydrocarbons, and other residual constituents including
oxygen, which method comprises passing the gases through
a reactor comprising a bed of active material (320) in an
enclosure having gas flow conduits (301,302) for
directing gas flow through or over the bed of active
10 material (320), applying an electrical potential to
generate a non-thermal plasma in gas permeating the
active material, at least a component of the active
material (320) being such as to adsorb or trap
carbonaceous particulates including soot, characterised
15 in that the electrical potential is applied to generate
said non-thermal plasma during passage through the active
material of the gases undergoing treatment whereby the
trapped carbonaceous particulates including soot have a
longer effective residence time in the non-thermal plasma
20 relative to species in the gas flow which are not
adsorbed or trapped and are oxidised by oxidative species
present in the gases while conversion of NO to NO₂ is
much less likely to occur.
- 25 2. A method as claimed in claim 1, further
characterised in that the gases are further subjected to
the action of a NO selective catalyst which selectively
absorbs both NO and hydrocarbons and/or partially
oxygenated hydrocarbons and promotes their reaction
30 together to reduce NO directly to N₂.
3. A method as claimed in claim 2, further
characterised in that the gases subjected to the action
of a NO selective catalyst are also subjected to further
35 plasma activation which promotes the formation of

activated hydrocarbons and/or partially oxygenated hydrocarbons.

4. A method as claimed in claim 2 or claim 3, further
5 characterised in that the NO selective catalyst comprises silver doped alumina.

5. A method as claimed in any of the preceding claims,
further characterised in that the bed of active material
10 (320) comprises beads of alumina.

6. A method as claimed in any of the preceding claims,
further characterised in that the bed of active material
includes a combustion catalyst.

15

7. A method as claimed in claim 6, further
characterised in that the combustion catalyst comprises
one or more materials from the group comprising alkali
metal oxide, or lanthanum oxide/alkali metal
20 oxide/vanadium pentoxide, vanadates such as metavanadates
and pyrovanadates.

8. A method as claimed in claim 6 or claim 7, further
characterised in that oxidation to carbon dioxide and
25 carbon monoxide of the said carbonaceous particulates
including soot occurs at temperatures lower than the
respective thermal oxidation temperature thereof.

9. A method as claimed in claim 8, further
30 characterised in that the said oxidation to carbon
dioxide and carbon monoxide occurs at temperatures lower
than 250°C.

10. A method as claimed in claim 8, further
35 characterised in that the said oxidation to carbon

dioxide and carbon monoxide occurs at temperatures as low as 100°C.

11. A method as claimed in any of the preceding claims,
5 further characterised in that the gases are subjected to flow through or over a plurality of beds of active material each of which adsorbs or traps a different predetermined chemical species.
- 10 12 A method as claimed in claim 11, further characterised in that in the plurality of beds of active material there is included, in addition to the active material for trapping carbonaceous particulates including soot, active material which adsorbs or traps one or more
15 predetermined chemical species from the group comprising nitrogen, oxygen, oxides of carbon such as CO, CO₂, water, hydrocarbons including saturated, unsaturated, cyclic, branched and un-branched hydrocarbons, oxygenated hydrocarbons such as aldehydes, ketones, alcohols, acids
20 ethers and esters, aromatic hydrocarbons and derivatives thereof including poly aromatic hydrocarbon compounds, oil fractions, fuel and partially burned fuel, air and air/fuel mixes, sulphur compounds including SO₂ and sulphates, organo-nitrogen species, acid gases,
25 combustion modifiers/enhancers, additives such as urea, ammonia, cerium oxide (such as Eolys) and plasma activated species such as O, OH, O₃ activated hydrocarbons including partially oxygenated hydrocarbons/organic molecules and electronically and
30 vibrationally excited state species.
13. A method as claimed in claim 12, further characterised in that the said additional active material is appropriately selected from the group comprising

dielectric or ferroelectric material, polymeric material, and ceramic material.

14. A method as claimed in any of the preceding claims, further characterised in that the bed or beds of active
5 material (320) is provided in the form of sheets, wafers, meshes, frits, coils, spheres, pellets, extrudate, granules, fibres, foams or honeycomb monolith or as a coating on sheets, wafers, meshes, frits, coils, spheres, pellets, extrudates, granules, fibres or honeycomb
10 monolith, foam, or membrane.

15. A method as claimed in any of the preceding claims, further characterised in that dielectric barrier material (304) between the electrodes (303,310) establishes a
15 dielectric barrier discharge type of reactor.

16. A non-thermal plasma reactor for the treatment of gases, which contain nitrogen oxides, carbonaceous particulates including soot, hydrocarbons, and other
20 residual constituents including oxygen, which reactor comprises a bed of active material (320) in an enclosure having gas flow conduits (301,302) for directing gas to flow through or over the bed of active material (320), electrodes (303,310) adapted when electrically energised
25 to generate non-thermal plasma in the gas permeating the active material (320), at least a component of the active material (320) being such as to adsorb or trap carbonaceous particulates including soot in the gas flow, characterised in that in operation of the reactor said
30 active material (320) increases the effective residence time in the non-thermal plasma of the said carbonaceous particulates including soot relative to the residence time of species in the gas flow which are not adsorbed or trapped, and the trapped carbonaceous particulates
35 including soot are oxidised by oxidative species present

- in the gases while conversion of NO to NO₂ is much less likely to occur, and an NO selective catalyst is additionally provided for selectively adsorbing both NO and hydrocarbons and/or partially oxygenated hydrocarbons, and promoting their reaction together to reduce NO directly to N₂.
17. A non-thermal plasma reactor as claimed in claim 16, further characterised in that the NO selective catalyst comprises silver doped alumina.
18. A non-thermal plasma reactor as claimed in claim 16 or claim 17, further characterised in that the bed of active material (320) comprises beads of alumina.
19. A non-thermal plasma reactor as claimed in any of claims 16 to 18, further characterised in that the bed of active material includes a combustion catalyst.
20. A method as claimed in claim 19, further characterised in that the combustion catalyst comprises one or more materials from the group comprising alkali metal oxide, or lanthanum oxide/alkali metal oxide/vanadium pentoxide, vanadates such as metavanadates and pyrovanadates.

INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference 15438 LgCm	FOR FURTHER ACTION See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/GB00/03943	International filing date (day/month/year) 16/10/2000	Priority date (day/month/year) 22/10/1999
International Patent Classification (IPC) or national classification and IPC B01D53/32		

Applicant

[AEA TECHNOLOGY PLC et al.]


"Accoratus PLC"

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of 7 sheets, including this cover sheet.
- ☐ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☐ Certain defects in the international application
- VIII ☒ Certain observations on the international application

Date of submission of the demand 02/02/2001	Date of completion of this report 23.01.2002
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epimu d Fax: +49 89 2399 - 4465	Authorized officer Connor, M Telephone No. +49 89 2399 8402



**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT**

International application No. PCT/GB00/03943

I. Basis of the report

1. With regard to the elements of the international application (*Replacement sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to this report since they do not contain amendments (Rules 70.16 and 70.17)*):

Description, pages:

1-16 as originally filed

Claims, No.:

1-22 as originally filed

Drawings, sheets:

1/1 as originally filed

2. With regard to the **language**, all the elements marked above were available or furnished to this Authority in the language in which the international application was filed, unless otherwise indicated under this item.

These elements were available or furnished to this Authority in the following language: , which is:

- ☐ the language of a translation furnished for the purposes of the international search (under Rule 23.1(b)).
- ☐ the language of publication of the international application (under Rule 48.3(b)).
- ☐ the language of a translation furnished for the purposes of international preliminary examination (under Rule 55.2 and/or 55.3).

3. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international preliminary examination was carried out on the basis of the sequence listing:

- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ The statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ The statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

4. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:

**INTERNATIONAL PRELIMINARY
EXAMINATION REPORT**

International application No. PCT/GB00/03943

☐ the drawings, sheets:

5. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

(Any replacement sheet containing such amendments must be referred to under item 1 and annexed to this report.)

6. Additional observations, if necessary:

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes:	Claims
	No:	Claims 1,13,14
Inventive step (IS)	Yes:	Claims
	No:	Claims 1-22
Industrial applicability (IA)	Yes:	Claims 1-22
	No:	Claims

2. Citations and explanations
see separate sheet

VIII. Certain observations on the international application

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:
see separate sheet

Re Item V**Reasoned statement under Rule 66.2(a)(ii) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

1.1 The present application concerns a non-thermal plasma reactor (claims 13 and 14) and a method (claim 1) of manufacturing a component thereof. The non-thermal reactor of claim 14 comprises:

- (a) an enclosure having;
 - (a1) gas flow conduits (301, 302) for directing the flow through or over the bed (320) (cf. (b));
 - (a2) electrodes (303, 310) adapted to generate non-thermal plasma in the gas;
- (b) said enclosure containing a bed of active material (320);
 - (b1) at least a component of which being modified so as
 - (b1i) to trap (e.g., adsorb) a predetermined chemical species in the gas flow;
 - (b1ii) to increase the residence time of the said species compared with the one of species of the gas flow which are not trapped; and
 - (b1iii) so modify the gas phase and/or surface chemistry of the treatment.

Method claim 1 consists of assembling (b) into (a) (which has (a1) and (a2)) and selecting a component (b1) of (b) for its capability to fulfil (b1i)-(b1iii). Product claim 13 is recited as a "product by process" claim referring back to process claim 1 and dependent claims thereof.

1.2 The claimed invention is said to have the advantage of allowing to select the residence time of various specific species composing a gas flow passing through a non-thermal plasma reactor, independently of gas flow rate, by trapping them into e.g. an adsorbent. Specific species requiring longer plasma treatment times (at a given power) than other species of the gas flow can thus be exposed to the plasma for a longer time, while the gas flow rate remains unaffected.

2 Reference is made to the following documents:

- D1: US-A-5 746 984 (HOARD JOHN W) 5 May 1998 (1998-05-05)
- D2: PATENT ABSTRACTS OF JAPAN vol. 1999, no. 09, 30 July 1999 (1999-07-30) & JP 11 114351 A (SANDENSHA:KK;YAMAMOTO TOSHIKI; YOU SADAYOSHI), 27 April 1999 (1999-04-27)
- D3: US-A-5 715 677 (VOGTLIN GEORGE E ET AL) 10 February 1998 (1998-02-10)
- D4: PATENT ABSTRACTS OF JAPAN vol. 1999, no. 09, 30 July 1999 (1999-07-30) & JP 11 114359 A (AGENCY OF IND SCIENCE &TECHNOL), 27 April 1999 (1999-04-27)

3 The method proposed in **claim 1** and the product in **claims 13** and **14** of the present application cannot be considered as novel (Article 33(2) PCT) for the following reasons.

- 3.1 In the following, claim 14 is addressed more specifically. Method claim 1 consisting merely in selecting and assembling the features called for in claim 14, and product claim 13, 'formally' depending on claim 1 are thus subject to the same objections as raised against claim 14. In present point 3, when "claim 14" is mentioned, it therefore refers to claims 1, 13, and 14.
- 3.2 All the features called for in claim 14 of the present application are anticipated in combination by each of D1-D4, as demonstrated in Table 1 below. It can be seen that all the apparatus features are disclosed in each of D1-D4, including the features relating to the use of the apparatus (features b1i-b1iii) (cf. point VIII.1 below). Consequently, the reactor *per se* (i.e., independently of its use) and the method of manufacturing it cannot be considered as novel in the sense of Article 33(2) PCT.
- 3.3 For applicant's information in case it decided to proceed in a national or regional phase, it is observed that no claim calls for a method for the treatment of gases by a non-thermal plasma as disclosed in the present description. It cannot be assessed from the abstract of the Japanese documents D2 and D4 whether they anticipate or not said method. D1 and D3, however, disclose a different treatment method where adsorption and plasma treatment stages are run separately. In case the applicant would file a claim directed to a method of non-thermal plasma treatment of gases, it is requested to file therewith a discussion on novelty and inventive step ("problem-solution" approach) with regard to D2 and D4 (if considered necessary, a translation of a part or the whole of D2 and D4 could be filed as well).

Table 1: features of claim 14 anticipated by D1-D4

feature	D1	D2	D3	D4
(a)	Figure 2: element #3 and Figure 4	Figure: #31 ("reactor body")	c.4, l.12: "housing 10 and 10' "	Figure: #9
...(a1)	Figure 4	Figure: #35, 38	Figure 1: #15, 15', 16, and 16'	Figure: gas flow path: a-b
...(a2)	Figure 4	Figure: #38, 39	c.4, l. 14: "a pair of electrodes 12-13"	Figure: #1, 2
(b)	Figure 4: electrified packed bed	Figure: #32 ("γ-alumina pellets")	c.4, l.13: "bed of granular absorbent/ catalyst material"	Figure: #3, 4
...(b1)	Figure 2: element #3, 5; c.5, l. 56-58	"adsorbent 32"	c.4, l.13: "absorbent/ catalyst material"	"porous adsorbent 3"
.....(b1i)*	c.3, l. 45-47 & c.5, l. 56	"waste gas adsorbed between the adsorbent"	c.4, l.13: "absorbent [...] material"	"porous adsorbent 3"
.....(b1ii)*	implicit: the non trapped species are not treated with plasma: c.3, l. 50	implicit: species being adsorbed are maintained within the plasma, whilst the non adsorbed species have left the reactor already.	implicit: the non trapped species are not treated with plasma: c.3, l. 7-30	implicit (cf. comment for D2)
.....(b1iii)*	implicit: adsorption & plasma treatment invariably modify the gas phase	implicit: the gas is separated by adsorption and modified by non thermal plasma.	implicit: adsorption & plasma treatment invariably modify the gas phase	implicit (cf. comment for D2)

* features relating to the use of the product called for in claim 14 (cf. point VIII.1 below)

- 4 Dependent claims 2-12 and 15-22 do not seem to contain any features which, in combination with the features of any claim to which they refer, meet the requirements of the PCT in respect of novelty or inventive step.

Re Item VIII

Certain observations on the international application

- 1 Features (b1i-b1iii) as disclosed in point V-1.1 *supra* lack clarity (Article 6 PCT) because they relate to the entity's use and not to the entity *per se*.

- 2 Claims 13 and 14 lack conciseness (article 6 PCT) as they effectively refer to the same invention. The applicant is requested to chose one among the two (preferably, claim 14 which defines the entity in terms of features thereof). If the applicant contests this view, its attention is drawn to the unity requirement set forth in Rule 13 PCT.
- 3 Claim 13 calls for a "reactor" referring back to the method of claims 1-12. Claims 1-12, however, address "a method of manufacturing a **component** for a [...] reactor" (bold added) and not a reactor. There is therefore a lack of clarity between the subject matter of claims 1 and 13 in contradiction with Article 6 PCT.

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 B01D53/32 F01N3/08 F01N3/01 F01N3/027

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 F01N B01D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

WPI Data, PAJ, EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 746 984 A (HOARD JOHN W) 5 May 1998 (1998-05-05) column 5, line 53 -column 8, line 61; figures 1-4	1-6, 8-16, 18, 19
X	PATENT ABSTRACTS OF JAPAN vol. 1999, no. 09, 30 July 1999 (1999-07-30) & JP 11 114351 A (SANDENSHA:KK;YAMAMOTO TOSHIKI; YOU SADAYOSHI), 27 April 1999 (1999-04-27) abstract	1, 13, 14, 18
A	US 5 715 677 A (VOGT LIN GEORGE E ET AL) 10 February 1998 (1998-02-10) abstract; figures	1, 14, 20-22
	-/-	

☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

* Special categories of cited documents:

- *A* document defining the general state of the art which is not considered to be of particular relevance
- *E* earlier document but published on or after the international filing date
- *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- *O* document referring to an oral disclosure, use, exhibition or other means
- *P* document published prior to the international filing date but later than the priority date claimed

T later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

X document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

Y document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

& document member of the same patent family

Date of the actual completion of the international search

9 January 2001

Date of mailing of the international search report

15/01/2001

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,
Fax (+31-70) 340-3016

Authorized officer

Sideris, M

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>PATENT ABSTRACTS OF JAPAN vol. 1999, no. 09, 30 July 1999 (1999-07-30) & JP 11 114359 A (AGENCY OF IND SCIENCE & TECHNOL), 27 April 1999 (1999-04-27) abstract</p> <hr/>	

Patent document cited in search report		Publication date	Patent family member(s)		Publication date
US 5746984	A	05-05-1998	AU	3403397 A	21-01-1998
			WO	9800221 A	08-01-1998
JP 11114351	A	27-04-1999	NONE		
US 5715677	A	10-02-1998	NONE		
JP 11114359	A	27-04-1999	NONE		

PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference 15438 LgCm	FOR FURTHER ACTION see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. PCT/GB 00/ 03943	International filing date (day/month/year) 16/10/2000	(Earliest) Priority Date (day/month/year) 22/10/1999
Applicant AEA TECHNOLOGY PLC et al.		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 4 sheets.



It is also accompanied by a copy of each prior art document cited in this report.

1. Basis of the report

- a. With regard to the **language**, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.



the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

- b. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international search was carried out on the basis of the sequence listing :



contained in the international application in written form.



filed together with the international application in computer readable form.



furnished subsequently to this Authority in written form.



furnished subsequently to this Authority in computer readable form.



the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.



the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

2. ☐ **Certain claims were found unsearchable** (See Box I).

3. ☐ **Unity of invention is lacking** (see Box II).

4. With regard to the **title**,



the text is approved as submitted by the applicant.



the text has been established by this Authority to read as follows:

5. With regard to the **abstract**,



the text is approved as submitted by the applicant.



the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the **drawings** to be published with the abstract is Figure No.



as suggested by the applicant.



because the applicant failed to suggest a figure.



because this figure better characterizes the invention.

1



None of the figures.

Box III TEXT OF THE ABSTRACT (Continuation of item 5 of the first sheet)

- line 1, add "(300)" after "reactor"
- line 2, add "(320)" after "material"

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 B01D53/32 F01N3/08 F01N3/01 F01N3/027

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 F01N B01D

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

WPI Data, PAJ, EPO-Internal

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 5 746 984 A (HOARD JOHN W) 5 May 1998 (1998-05-05) column 5, line 53 -column 8, line 61; figures 1-4	1-6, 8-16, 18, 19
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	--- -/--	

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* Special categories of cited documents:

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- *O* document referring to an oral disclosure, use, exhibition or other means
- *P* document published prior to the international filing date but later than the priority date claimed

- *T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- *X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- *Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- *&* document member of the same patent family

Date of the actual completion of the international search

9 January 2001

Date of mailing of the international search report

15/01/2001

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,
Fax: (+31-70) 340-3016

Authorized officer

Sideris, M

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	<p>PATENT ABSTRACTS OF JAPAN vol. 1999, no. 09, 30 July 1999 (1999-07-30) & JP 11 114359 A (AGENCY OF IND SCIENCE &AMP;TECHNOL), 27 April 1999 (1999-04-27) abstract</p> <p>-----</p>	

Patent document cited in search report		Publication date	Patent family member(s)		Publication date
US 5746984	A	05-05-1998	AU	3403397 A	21-01-1998
			WO	9800221 A	08-01-1998
JP 11114351	A	27-04-1999	NONE		
US 5715677	A	10-02-1998	NONE		
JP 11114359	A	27-04-1999	NONE		